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Rigorous Bounds and Relations among Spatial and Temporal Approximations in the Theory of Combustion

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Abstract—Rigorous bounds on temperature and concentration are obtained for a model reacting system incorporating spatial variation, reactant consumption, and Arrhenius kinetics. Similar bounds are obtained for the Semenov (spatially uniform) approximation. Temperatures predicted by the Frank-Kamenetskii (stationary) approximation are shown to be upper bounds on the temperature of the complete time-dependent system. The latter supports numerical and approximate analytic arguments that the critical value of the Frank-Kamenetskii heat generation parameter in the stationary approximation is a lower bound on analogously defined critical parameter values in the complete system. The fundamental mathematical tool is a comparison result for systems of parabolic partial differential equations.

A comprehensive model for studying thermal explosion phenomenon that incorporates both temporal and spatial variations in temperature and reactant density is given by the initial-boundary-value problem

$$\rho C_1 T_t^* = \kappa \nabla^2 T^* + Q A c^m e^{-E/RT^*}, \quad (1)$$

$$c_t^* = \mu \nabla^2 c^* - A c^m e^{-E/RT^*}, \quad (2)$$

$$\kappa T_{\nu}^* = \chi(T - T_a) = 0, \quad c_{\nu}^* = 0, \quad x^* \in \partial D, \quad t^* = 0, \quad (3)$$

$$T(x^*, 0) = T_a, \quad c(x^*, 0) = c_0, \quad x^* \in D, \quad (4)$$

(Symbols are defined below in Nomenclature. Subscripts denote partial derivatives; for example, $T_t^* = \partial T / \partial t^*$. The independent variable x^* may be a scalar if D is a one-dimensional domain or a two- or three-dimensional vector if D is a plane region or a solid.)

Because of the essential nonlinearities and the coupling, it is formidable to analyze the above system in its present form. To make the problem tractable, it is customary to make various approximations. The two commonly encountered simplifications are the Semenov and Frank-Kamenetskii approximations. In the former case, spatial variation is completely neglected. The thermal

dissipation by diffusion through the solid reactant is replaced by Newtonian cooling at the boundary of a solid whose temperature is assumed spatially uniform:

$$\rho C_1 VT'(t^*) = \frac{\chi S(T - T_a)}{T_a} + Q A V c^m e^{-E/RT}, \quad (5)$$

$$c'(t^*) = A c^m e^{-E/RT}, \quad c(0) = c_0, \quad (6)$$

In the second approximation, one ignores reactant consumption while retaining thermal diffusion in a so-called stationary approximation:

$$0 = \kappa \nabla^2 T^* + Q A e^{-E/RT} \quad (7)$$

$$\kappa T_{\nu}^* = \chi(T - T_a) = 0, \quad x^* \in \partial D, \quad (8)$$

Many investigators have studied these approximations, usually by imposing special conditions on the parameters or by simplifying nonlinear terms; for examples, see (Gray and Lee, 1967a), (Merzhanov and Averson, 1971). Fewer have explored relationships among these approximations and the complete problem (1-4); a recent study that emphasizes connections between parameter groups in the various approximations, especially when the kinetics term $e^{-E/RT}$ is simplified, is that of Boddington, Gray, and Wake (1977).

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Here we shall take a somewhat different approach to examining relationships among these approximations and the complete problem. By constructing solutions of appropriate differential inequalities, we can, for example, rigorously establish that the temperatures in (1-4) or (5-6) are bounded for all time or that the temperature field defined by the stationary approximation (7-8) is larger at every point of the reacting solid than the temperature field of the complete problem (1-4), regardless of the time. We emphasize that these bounds are mathematically rigorous and that they involve no special restrictions on parameters, approximations to kinetics, or the like.

To explain the physical idea behind such bounds, suppose that the history of reactant consumption at an extremely low temperature, say that of the surrounding medium, is available. Since reactant cannot be consumed more slowly, this curve of reactant density against time must be an upper bound on the time course of density in the actual reaction. If the reaction could now be repeated with reactant being available in amounts dictated by this upper bound curve, the temperature of the solid in this approximate reaction would be above that in the actual reaction because more of the exothermic reactant would be available at any given time. In other words, underestimating temperature would overestimate reactant as a function of time. Overestimating reactant would then overestimate temperature as a function of time.

After the introduction of a set of dimensionless variables in Section 2, we derive the rigorous bounds on the dimensionless temperature and concentration. The relation of the complete problem (1-4) to the approximate problems of Semenov and Frank-Kamenetskii is then considered in Sections 4 and 5, respectively. For the sake of simplicity of exposition, we have considered here a model for a single species. The techniques developed in this paper, however, are capable of handling multiple species. Also, the lack of a diffusion term in Eq. (2) ($\mu^* = 0$) causes no significant changes.

2 DIMENSIONLESS VARIABLES

To nondimensionalize the complete problem (1-4) and Frank-Kamenetskii's stationary approximation (7-8), we introduce the usual temperature, concentration, and length scales; see θ , λ , and x in

Nomenclature. As a time scale, we use the characteristic reaction time,

$$t_R^* \equiv eE RT_0 / Ac_0^m \quad (1)$$

the time required to consume an initial concentration of c_0 in a spatially uniform reaction proceeding at the (constant, initial) ambient temperature rate; i.e., $1/t_R^*$ is the initial reaction rate. We obtain the non-dimensionalized complete problem, denoted by (CP):

$$\sigma_D \theta_t - \nabla^2 \theta = \delta \lambda^m f(\theta) \quad (\text{CP-1})$$

$$\lambda_t = \mu \nabla^2 \lambda - \lambda^m f(\theta) \quad (\text{CP-2})$$

$$\theta_\nu \cdot \mathbf{B}i \theta = 0, \quad \lambda_\nu = 0, \quad x \in \partial D, \quad t \geq 0, \quad (\text{CP-3})$$

$$\theta(x, 0) = 0, \quad \lambda(x, 0) = 1, \quad x \in \partial D. \quad (\text{CP-4})$$

Proceeding as above but writing ψ_ϵ for the scaled temperature, we obtain the non-dimensionalized Frank-Kamenetskii stationary approximation, denoted by (FK $_\epsilon$):

$$0 = \nabla^2 \psi_\epsilon - \delta f(\psi_\epsilon) \quad (\text{FK}_\epsilon-1)$$

$$\psi_{\epsilon, \nu} \cdot \mathbf{B}i \psi_\epsilon = 0, \quad \psi_\epsilon \in \partial D. \quad (\text{FK}_\epsilon-2)$$

(If $\epsilon = 0$, then $f(\psi) = e^\psi$ in (FK $_0$ -1). The distinction between the cases $\epsilon = 0$ and $\epsilon > 0$ in the kinetics term will be important later.) The meaning of ψ_ϵ when (FK $_\epsilon$) exhibits multiple solutions will be clarified in Section 5.

The parameter σ_D is the ratio of a characteristic diffusion time $t_D^* \equiv \rho C_1 d^2 / \kappa$ to the characteristic reaction time t_R^* ; that is, σ_D is a non-dimensionalized characteristic diffusion time. Alternatively, σ_D is a Damköhler number, the ratio of reaction rate at $\theta = 0$ to the rate of thermal diffusion. Of course, δ is the usual Frank-Kamenetskii parameter, a non-dimensionalized heating rate or a non-dimensionalized temperature increase when diffusion is the only heat dissipation mechanism.

With the same choice of temperature, reactant, and time scales, we obtain from (5-6) the non-dimensionalized Semenov approximation, denoted by (SA),

$$\sigma_N \theta'(t) - \theta = a \lambda^m f(\theta), \quad \theta(0) = 0, \quad (\text{SA-1})$$

$$\lambda'(t) = -\lambda^m f(\theta), \quad \lambda(0) = 1, \quad (\text{SA-2})$$

The parameters σ_N and α are analogous to σ_D and δ in (CP): $\sigma_N \equiv t_N^*/t_R^*$ is a non-dimensionalized characteristic time for Newtonian cooling, and α is a non-dimensionalized temperature increase when Newtonian cooling is the only heat dissipation mechanism. To define t_N^* , suppose $T(t)$ satisfies (5) with $Q = 0$. If $T(0) - T_a > 0$, then choose t_N^* so that $T(t_N^*) - T_a = (T(0) - T_a)/e$.

Note that $a/(V/\chi S) = \delta/(d^2/\kappa)$. Since the parameter groups $V/\chi S$ and d^2/κ are volume specific heat loss terms for the Semenov approximation and complete problems, respectively, we see that the heat generation term in each problem has been scaled against the appropriate dissipation factor.

If the dissipation terms $\nabla^2\theta$ in (CP-1) and $-\theta$ in (SA-1) are eliminated, then dividing (CP-2) into (CP-1) and (SA-2) into (SA-1) yields, respectively, $d\theta/d\lambda = -\delta/\sigma_D$ for (CP) and $d\theta/d\lambda = -\alpha/\sigma_N$ for (SA). These quotients are equal:

$$\delta/\sigma_D = \alpha/\sigma_N \equiv B.$$

Since $B = -d\theta/d\lambda$, which is the temperature increase per mole of reactant consumed in the absence of heat loss, B is the non-dimensionalized adiabatic temperature rise.

One of the aims of this paper is exploring the effects of approximations of the highly nonlinear reaction terms, specifically the exponential approximation. In our notation, the exponential approximation corresponds to setting $\epsilon = 0$ in the expression for $f(\theta)$. For example, in the case of the Frank-Kemenetskii stationary approximation, $(F-K_0)$ denotes setting $\epsilon = 0$ in the kinetics term in $(F-K_\epsilon)$, thereby replacing $f(\theta) = \exp(\theta/(1+\epsilon\theta))$ by e^θ . References to $\epsilon = 0$ or to infinite activation energy in the following are to the *kinetics term alone*, not to any other term or parameter appearing in these nondimensionalized equations. Readers accustomed to asymptotic analyses of these models for small ϵ (large activation energy) should be especially alert to our treatment of ϵ as an independent parameter in the kinetics terms of the dimensionless models.

3 RIGOROUS BOUNDS

Here we shall develop a technique for rigorously confirming the bounds on $\theta(x, t)$ and $\lambda(x, t)$ suggested by the intuitive arguments in the Introduction. We shall exhibit systems of differential inequalities which are bounding problems in the

sense that any pair of functions satisfying such a system of inequalities is a bound on the pair $(\theta(x, t), \lambda(x, t))$.

To summarize the comparison theorem of (Chandra and Davis, 1979), which is central to these arguments, consider a given initial-boundary-value problem for a system of parabolic differential equations:

$$u_t(x, t) = k_1 \nabla^2 u + F(u, v) \quad (9)$$

$$v_t(x, t) = k_2 \nabla^2 v + G(u, v) \quad (10)$$

$$a_1 u_p + b_1 u = 0, \quad a_2 v_p + b_2 v = 0, \quad x \in \partial D, \quad t > 0, \quad (11)$$

$$u(x, 0) = u_0(x), \quad v(x, 0) = v_0(x), \quad x \in D. \quad (12)$$

We suppose that $k_1 \geq 0$, $k_2 \geq 0$ (to guarantee the system is at least weakly parabolic) and that $a_i, b_i \geq 0$, $a_i^2 + b_i^2 > 0$ if $k_i > 0$ while $a_i = b_i = 0$ if $k_i = 0$ (to guarantee the problem is neither over- nor under-determined by properly posed boundary conditions). We also assume each smooth solution (u, v) depends continuously upon the parameters in the differential equations and upon the initial and boundary data. We make no restrictions on the monotonicity of the given nonlinear terms F and G .

However, we assume there are functions F and G with the properties

$$(i) \quad F(u, v) \leq F(u, v), \quad G(u, v) \leq G(u, v),$$

$$(ii) \quad F(u, v) \text{ is monotone nondecreasing in } v \text{ for each fixed } u \text{ (i.e., } v \leq w \text{ implies } F(u, v) \leq F(u, w) \text{ for each } u) \text{ and } G(u, v) \text{ is monotone nondecreasing in } u \text{ for each fixed } v.$$

(Property (ii) which is essentially monotonicity in the off-diagonal variables of F and G , is known as quasi-monotonicity in the mathematical literature.)

Finally, we suppose that $(u(x, t), v(x, t))$ is any pair of functions satisfying the following system of differential inequalities

$$u_t(x, t) \leq k_1 \nabla^2 u + F(u, v) \quad (13)$$

$$v_t(x, t) \leq k_2 \nabla^2 v + G(u, v) \quad (14)$$

$$a_1 u_p + b_1 u \leq 0, \quad a_2 v_p + b_2 v \leq 0, \quad x \in \partial D, \quad t > 0, \quad (15)$$

$$u(x, 0) \leq u_0(x), \quad v(x, 0) \leq v_0(x), \quad x \in D. \quad (16)$$

Comparison Result: If (u, v) is the solution of (9-12) and $(\underline{u}, \underline{v})$ is any pair of functions satisfying (13-16), then $(\underline{u}, \underline{v})$ is a lower bound on (u, v) ; i.e., $(\underline{u}, \underline{v}) \leq (u, v)$ (meaning both $\underline{u}(x, t) \leq u(x, t)$ and $\underline{v}(x, t) \leq v(x, t)$ for all $x \in D$ and all $t \geq 0$.) If F and G in (13) and (14) are replaced by functions satisfying Property (i) with all inequalities reversed and Property (ii), then solutions of (13-16) with all inequalities reversed are *upper* bounds on (u, v) .

We shall repeatedly use this Comparison Result by constructing appropriate quasi-monotone nonlinearities derived from the kinetics terms of the combustion problems in question and by studying various pairs of bounding functions satisfying the appropriate version of (13-16).

Now consider the particular problem (CP), in which $u = \theta, v = \lambda, k_1 = 1, k_2 = \mu, F = \delta \lambda^m f(\theta), G = \lambda^m f(\theta)$, and so on. To construct a lower bounding problem whose nonlinear terms satisfy Properties (i) and (ii), we define

$$F(\theta, \lambda) = \inf\{F(\theta, z); \lambda \leq z\},$$

$$G(\theta, \lambda) = \inf\{G(z, \lambda); \theta \leq z\},$$

where "inf" abbreviates infimum, or greatest lower bound. By definition, $F \leq F$ and $G \leq G$. The quasi-monotonicity condition is automatically satisfied: F is nondecreasing in λ (because the infimum is taken over smaller sets as λ increases) and G is nondecreasing in θ . We find that $F(\theta, \lambda) = F(\theta, \lambda) = \delta \lambda^m f(\theta)$ and $G(\theta, \lambda) = \lambda^m f(\theta)$.

Let $(\underline{\theta}, \underline{\lambda})$ be a pair of functions which satisfy

$$\sigma_D \underline{\theta}_t - \nabla^2 \underline{\theta} = \delta \underline{\lambda}^m f(\underline{\theta}) \quad (17)$$

$$\underline{\lambda}_t - \mu \nabla^2 \underline{\lambda} = c^{-1} \underline{\lambda}^m \quad (18)$$

$$\underline{\theta}_\nu \leq Bi \underline{\theta} = 0, \underline{\lambda}_\nu = 0, x \in \partial D, t \geq 0, \quad (19)$$

$$\underline{\theta}(x, 0) = 0, \underline{\lambda}(x, 0) = 1, x \in D. \quad (20)$$

According to the Comparison Result, (17-20) is a lower bounding problem for (CP) in the sense that $(\underline{\theta}, \underline{\lambda}) \leq (\theta(x, t), \lambda(x, t))$ for any pair (θ, λ) which satisfies (17-20).

Intuitively, this assertion is reasonable because (17-20) models an approximate reaction which starts with less, colder reactant (cf. (20) and (CP-4)) and proceeds to completion in a colder environment into which there is a greater flux of heat (cf. (19) and (CP-3)). Reactant is consumed

in (18) as if the reaction were proceeding at infinite temperature ($f(\tau) = c^{-1}\tau$). Heat is released in (17) by the same mechanism as in the actual reaction, but less reactant is available in (17) at any given time because it is being consumed at a more rapid, higher temperature rate.

Note that $\underline{\theta} = 0, \underline{\lambda} = 0$ satisfy (17-20), thereby forcing $0 \leq \theta(x, t), 0 \leq \lambda(x, t)$.

With these physically obvious lower bounds in hand, we can now construct an upper bounding problem employing the minimum temperature $\underline{\theta} = 0$. Define

$$\bar{F}(\theta, \lambda) = \sup\{F(\theta, z); 0 \leq z \leq \lambda\}, \quad (21)$$

$$\bar{G}(\theta, \lambda) = \sup\{G(z, \lambda); 0 \leq z \leq \theta\}, \quad (22)$$

where "sup" abbreviates supremum, or least upper bound. We find that $\bar{F}(\theta, \lambda) = F(\theta, \lambda) = \delta \lambda^m f(\theta)$ and $\bar{G}(\theta, \lambda) = \lambda^m f(\theta) = \lambda^m$.

Let $(\bar{\theta}, \bar{\lambda})$ be a pair of functions which satisfy

$$\sigma_D \bar{\theta}_t - \nabla^2 \bar{\theta} = \delta \bar{\lambda}^m f(\bar{\theta}) \quad (23)$$

$$\bar{\lambda}_t - \mu \nabla^2 \bar{\lambda} = \bar{\lambda}^m \quad (24)$$

$$\bar{\theta}_\nu = Bi \bar{\theta} = 0, \bar{\lambda}_\nu = 0, x \in \partial D, t \geq 0, \quad (25)$$

$$\bar{\theta}(x, 0) = 0, \bar{\lambda}(x, 0) = 1, x \in D. \quad (26)$$

This system represents an approximate reaction whose physical basis is the dual of (17-20). An analogous appeal to the Comparison Result demonstrates that (23-26) is an upper bounding problem for (CP) in the sense that $(\theta(x, t), \lambda(x, t)) \leq (\bar{\theta}, \bar{\lambda})$ for any pair $(\bar{\theta}, \bar{\lambda})$ satisfying (23-26).

Similar arguments permit the construction of bounding problems for (SA). The corresponding lower bounding problem is

$$\sigma_N \underline{\theta}'(t) = \underline{\theta} - \alpha \lambda^m(\underline{\theta}), \underline{\theta}(0) = 0, \quad (27)$$

$$\underline{\lambda}'(t) = c^{-1} \underline{\lambda}^m, \underline{\lambda}(0) = 1. \quad (28)$$

Once again, we may take $(\underline{\theta}, \underline{\lambda}) = (0, 0)$ to conclude $(0, 0) \leq (\theta(t), \lambda(t))$ for the solution $(\theta(t), \lambda(t))$ of (SA). Using (21-22), we then define an upper bounding problem for (SA):

$$\sigma_N \bar{\theta}'(t) = \bar{\theta} - \alpha \bar{\lambda}^m f(\bar{\theta}), \bar{\theta}(0) = 0, \quad (29)$$

$$\bar{\lambda}'(t) = \bar{\lambda}^m, \bar{\lambda}(0) = 1. \quad (30)$$

To exhibit a bound on the solution of (CP) or (SA), we need only find a pair of functions satisfying the appropriate bounding problem. Since these bounding problems are defined by differential inequalities, one may, in principle, construct a number of different bounding pairs from a single bounding problem.

4 BOUNDS FOR THE SEMENOV APPROXIMATION AND THE COMPLETE PROBLEM

Without making the assumption $\epsilon = 0$, we shall obtain spatially uniform bounds on temperature and reactant concentration for both (SA) and (CP) by studying a pair of ordinary differential equations whose solution satisfies an upper bounding problem for (SA) and (CP). Roughly, these bounds show that bounded kinetics (*i.e.*, finite activation energy or $\epsilon > 0$) imply bounded temperatures. When $\epsilon = 0$, in which case the kinetics term is unbounded due to infinite activation energy, these techniques provide a bound on induction time based on reactant consumption occurring at ambient temperature ($\theta = 0$).

Let $(\bar{\theta}(t), \bar{\lambda}(t))$ denote the solution of

$$\bar{\theta}'(t) = B\bar{\lambda}^m(t)f(\bar{\theta}(t)), \quad \bar{\theta}(0) = 0 \quad (31)$$

$$\bar{\lambda}'(t) = -\bar{\lambda}^m(t), \quad \bar{\lambda}(0) = 1. \quad (32)$$

If B denotes $\delta\sigma_B$, then $(\bar{\theta}(t), \bar{\lambda}(t))$ evidently satisfies (23-26), an upper bounding problem for (CP). If B denotes $\alpha\sigma_A$, then $(\bar{\theta}(t), \bar{\lambda}(t))$ satisfies (29-30), a corresponding upper bounding problem for (SA). That is, $(\bar{\theta}(t), \bar{\lambda}(t)) = (\theta, \lambda)$ whether (θ, λ) satisfies (SA) or (CP).

To obtain an expression for $\bar{\theta}(t)$, substitute (32) in (31), separate variables, and integrate:

$$\int_0^{\bar{\theta}(t)} du f(u) = B(1 - \lambda(t)). \quad (33)$$

Solving (32) independently yields

$$\bar{\lambda}(t) = e^{-(m-1)t}, \quad m \neq 1, \quad (34a)$$

$$\bar{\lambda}(t) = [1 - (m-1)t]^{1/(1-m)}, \quad m \neq 1, \quad (34b)$$

(When $0 < m < 1$ in (34b), $\bar{\lambda}(t)$ takes the form given for $0 < t < 1/(1-m)$ and $\bar{\lambda}(t) = 0$ for $1/(1-m) < t$.)

Since $\lim_{t \rightarrow \infty} \bar{\lambda}(t) = 0$, $\lim_{t \rightarrow \infty} \bar{\theta}(t) \equiv \bar{\theta}(\infty)$ must satisfy

$$\int_0^{\bar{\theta}(\infty)} du f(u) = B. \quad (35)$$

Since $\bar{\theta}'(t) > 0$ in (31), $\bar{\theta}(t)$ is increasing and $\bar{\theta}(t) \rightarrow \bar{\theta}(\infty)$ for all t . Hence, $\bar{\theta}(\infty)$ is an upper bound on the maximum temperature in either (SA) or (CP).

From (35), we see that this bound $\bar{\theta}(\infty)$ on the maximum temperature depends only the dimensionless adiabatic temperature rise B . Neither dissipation nor Newtonian cooling is included. The problem (31-32) defining $(\bar{\theta}, \bar{\lambda})$ is similarly restricted as well; (31) could be thought of as an approximation to (SA-1) or (CP-1) in which dissipation has been eliminated by discarding the term $-\theta$ or $-\nabla^2\theta$. Since the spatially uniform function $\bar{\theta}(t)$ satisfies the boundary condition (CP-3) with $Bi = 0$, $\bar{\theta}(t)$ may be thought of as the temperature of a completely adiabatic reaction.

The supply of reactant to this adiabatic reaction is governed by (32), which is the rate equation at $\theta = 0$. That is, this hypothetical adiabatic reaction has more reactant available at any time than does (SA) or (CP) because it loses reactant at the slowest, ambient temperature rate.

These remarks provide intuitive confirmation of the rigorous result that temperature and reactant concentration in either (SA) or (CP) are bounded above by $(\bar{\theta}(t), \bar{\lambda}(t))$ and that the maximum temperature in either case is bounded by $\bar{\theta}(\infty)$. Further, they warn us that $\bar{\theta}(\infty)$ may substantially overestimate the true maximum temperature.

To show that $\bar{\theta}(\infty)$ is always finite, note that $f(u) = f(\infty) = e^{1/\epsilon}$. Then (35) yields

$$B \geq \int_0^{\bar{\theta}(\infty)} e^{-1/\epsilon} du = e^{-1/\epsilon} \bar{\theta}(\infty),$$

or

$$\bar{\theta}(\infty) \leq B e^{1/\epsilon}. \quad (36)$$

Boddington, Gray, and Wake (1977; Eq. (14)) have obtained a similar upper bound, $B\sigma_A e^{1/\epsilon}$ in our notation, in the uniform temperature case without reactant consumption but with dissipation (*i.e.*, (SA-1) alone with $\lambda \equiv 1$). We now see that a bound of this type applies as well to both the

uniform temperature case with reactant consumption (SA), as one might expect on physical grounds, and to the distributed temperature case with reactant consumption (CP), where the physical relation to the uniform temperature case is perhaps less obvious.

Put simply, (36) implies that the temperature in both (SA) and (CP) remains finite for all time so long as the activation energy is finite ($\epsilon > 0$).

Unfortunately, (35) cannot provide tighter bounds on the maximum temperature. Since $f(u) \rightarrow 1$, (35) yields

$$B \leq \int_0^{\bar{\theta}(\infty)} du = \bar{\theta}(\infty).$$

In physical terms, a bounding problem that neglects all dissipation will exhibit maximum temperatures at least as great as the adiabatic temperature rise.

We may, however, use the upper bound (36) to define an improved lower bounding problem for either (SA) or (CP) by replacing $f(x) = e^{1/\epsilon}$ in (18) or (28) by $f(Be^{1/\epsilon})$. If $\underline{\lambda}(t)$ now satisfies

$$\underline{\lambda}'(t) = \underline{\lambda}^m(t)f(Be^{1/\epsilon}), \quad \underline{\lambda}(0) = 1,$$

then the pair $(0, \underline{\lambda}(t))$ satisfies either of these improved lower bounding problems. We calculate

$$\underline{\lambda}(t) = \exp[-f(Be^{1/\epsilon})t], \quad m = 1,$$

$$\underline{\lambda}(t) = [1 + (m-1)f(Be^{1/\epsilon})t]^{1/(1-m)}, \quad m \neq 1.$$

(If $0 < m < 1$ in the last expression, then $\underline{\lambda}(t)$ takes the form given for $0 < t < [(1-m)f(Be^{1/\epsilon})]^{-1}$ and $\underline{\lambda}(t) = 0$ for larger values of t .)

Hence, if $\lambda(t)$ is defined by (34) and (θ, λ) is a solution of either (SA) or (CP),

$$(0, \underline{\lambda}(t)) \leq (\theta, \lambda) \leq (\bar{\theta}(t), \bar{\lambda}(t)) \leq (Be^{1/\epsilon}, \bar{\lambda}(t)). \quad (37)$$

Since the dimensionless time t has been scaled against t_R^* , the characteristic reaction time, (37) suggests that, in dimensional time, the time constant for reactant consumption ranges between $1/f(Be^{1/\epsilon})t_R^*$ and $1/t_R^*$.

If we make the exponential approximation $f(\theta) \approx e^\theta$, then this unbounded kinetics term permits $\bar{\theta}(t)$ in the adiabatic bounding problem (31-32) to grow without bound. To calculate the

induction time \bar{t}_i at which $\bar{\theta} = \infty$, write e^θ in place of $f(u)$ in (33) and integrate:

$$1 - e^{-\bar{\theta}(t)} = B(1 - \bar{\lambda}(t)).$$

If $\bar{\theta}(\bar{t}_i) = \infty$, then $1 = B(1 - \bar{\lambda}(\bar{t}_i))$, and

$$\bar{t}_i = \bar{\lambda}^{-1}(1 - 1/B),$$

where $\bar{\lambda}^{-1}$ denotes the inverse of the function $\bar{\lambda}$ defined by (34). Specifically, we have

$$\bar{t}_i = \ln(1 - 1/B), \quad m = 1,$$

$$\bar{t}_i = \frac{(1 - 1/B)^{1/(m-1)} - 1}{m-1}, \quad m \neq 1.$$

(Note that $B > 1$; extremes are $4 < B < 1,000$ while a typical value for spontaneous ignition of wood is $B \approx 30$ (Tyler and Wesley, 1967), (Gray and Lee, 1967b).) In either case, an easy argument shows $\bar{t}_i > 1/B$ or, in dimensional variables,

$$\bar{t}_i^* \geq \bar{t}_i t_R^* \geq t_R^*/B \geq t_{ad}^*,$$

where t_{ad}^* is the induction time for the spatially uniform adiabatic reaction with infinite activation energy and no reactant consumption. Of course, an induction time such as \bar{t}_i^* computed with reactant consumption should be larger than t_{ad}^* , an induction time computed without reactant consumption, as illustrated.

If $\bar{\theta}(t)$ continues to denote the solution of (31) with $\epsilon = 0$, then our same bounding arguments can show that $\bar{\theta}(t)$ is an upper bound on θ in either (SA) or (CP) with $\epsilon > 0$ (or $\epsilon = 0$ as well). Hence, neither of these problems can exhibit infinite temperature before $\bar{\theta}(t) = \infty$. That is, the induction period t_i for either (SA) or (CP) at infinite activation energy must satisfy

$$t_i \geq \bar{t}_i \geq 1/B \geq t_{ad}.$$

Hence, \bar{t}_i is a lower bound on the induction time in either (SA) or (CP) with $\epsilon > 0$.

5 THE RELATION BETWEEN THE STATIONARY APPROXIMATION AND THE COMPLETE PROBLEM

The basic result of this section is the following: if $\bar{\lambda}(t)$ is defined by (34), then

$$(\psi_0(x), \bar{\lambda}(t)), (\psi_\varepsilon(x), \bar{\lambda}(t)), (\theta(x, t), \lambda(x, t)), \quad (38)$$

$$\delta_{crit} = \delta_\varepsilon = \delta_{CP},$$

where ψ_ε is the stationary temperature of (FK_ε) and θ is the exact temperature of (CP). If δ in (FK₀) is supracritical, then $\psi_0(x)$ does not exist and only the second inequality is valid.

In other words, either of the stationary temperatures $\psi_0(x)$ or $\psi_\varepsilon(x)$ bounds the true temperature $\theta(x, t)$ given by (CP).

The first inequality in (38) is a consequence of $e^0 \cdot f(\theta)$ and the scalar maximum principle (Protter and Weinberger, 1967, p.64). To repeat the argument in slightly different terms, the relation $e^0 \cdot f(\theta)$ makes (FK₀) an upper bounding problem for (FK_ε).

To establish the second inequality in (38), we first note that direct calculation in (34) shows $\bar{\lambda}(t) = 1$ while the maximum principle for elliptic boundary value problems (Protter and Weinberger, 1967, p.64) forces $\psi_\varepsilon(x) = 0$. The former yields

$$\sigma_D \psi_{\varepsilon,t}(x) \equiv 0 \quad \nabla^2 \psi_\varepsilon = \delta f(\psi_\varepsilon) = \nabla^2 \psi_\varepsilon = \delta \bar{\lambda}^m f(\psi_\varepsilon) \quad (39)$$

while the latter ensures that ψ_ε satisfies the initial condition (26) in the upper bounding problem.

Comparing (39) and the defining equation (32) for $\bar{\lambda}(t)$ with (23) and (24), respectively, shows that $(\psi_\varepsilon(x), \bar{\lambda}(t))$ is a solution of the upper bounding problem (23-26) for (CP). Hence, $(\psi_\varepsilon(x), \bar{\lambda}(t)) = (\theta(x, t), \lambda(x, t))$.

If δ is chosen so that (FK_ε), $\varepsilon = 0$, has several solutions of which only one is stable, then $\psi_\varepsilon(x)$ is the smallest non-negative stable solution satisfying (25), for stability is the manifestation of the continuous dependence condition of the Comparison Result (Chandra and Davis, 1979), upon which rests the validity of our bounding claims. If δ is in a range in which (FK_ε) has several stable solutions, then $\psi_\varepsilon(x)$ may denote any one of these, including, of course, the smallest.

Suppose δ_ε and δ_{CP} are "critical" values of δ in (FK_ε) and (CP), respectively, defined by some reasonable criteria involving, say, a sharply increased maximum temperature. (Recall that results of the previous section guarantee that the temperature in (FK_ε), $\varepsilon = 0$, or (CP) is never infinite.) Let δ_{crit} be the usual Frank-Kamenetskii critical value for (FK₀) "beyond which this stationary distribution ceases to be possible" (Frank-Kamenetskii, 1969, p.345). Then the bounds of (38) suggest

a phenomena observed in numerical computations by Tyler and Wesley (1967) and Parks (1961) and in various approximate analytic studies by Frank-Kamenetskii (1945) (corrected in (Gray and Lee, 1967b)), Gray and Lee (1967b), Thomas (1961), Boddington, Gray, and Wake (1977), *et al.* For a specific comparison in the spatially uniform case, see (Kassoy and Liñán, 1978, Eq. (5.15)). That is, the critical value of the Frank-Kamenetskii parameter appears to be a lower bound on critical values for (FK_ε) or (CP) defined by sharp increases in temperature.

Just as in the previous section, the upper bounds on temperature of (38) lead to new lower bounds on reactant concentration. However, these bounds exhibit spatial variation as well.

Define $(\underline{\theta}(x, t), \underline{\lambda}(x, t))$ by

$$\sigma_D \underline{\theta}_t = \nabla^2 \underline{\theta} = \delta \underline{\lambda}^m f(\underline{\theta}), \quad (40)$$

$$\lambda_t = \mu \nabla^2 \underline{\lambda} = \underline{\lambda}^m f(\psi_\varepsilon(x)), \quad (41)$$

$$\underline{\theta}(x, 0) = 0, \quad \underline{\lambda}(x, 0) = 1, \quad x \in D \quad (42)$$

$$\underline{\theta}_v = Bi \underline{\theta} = 0, \quad \underline{\lambda}_v = 0 \quad x \in \partial D, \quad t > 0, \quad (43)$$

Then $(\underline{\theta}, \underline{\lambda})$ evidently satisfies an improved lower bounding problem obtained from (17-20) by replacing $f(\tau) = e^{1-\tau}$ in (18) by $f(\psi_\varepsilon(x))$. Hence, $(\underline{\theta}(x, t), \underline{\lambda}(x, t)) = (\theta(x, t), \lambda(x, t))$, where $(\underline{\theta}, \underline{\lambda})$ satisfies (40-43).

For a given reactant geometry, one could, in principle, compute $\psi_\varepsilon(x)$ and solve (41-42) for $\underline{\lambda}(x, t)$. Then the expression for $\underline{\lambda}(x, t)$ could be used in (40) to obtain a single differential equation for a non-trivial, spatially varying lower bound on temperature. The expression for $\lambda(x, t)$ would also provide a spatially varying lower bound on the time constant for reactant consumption; see the remarks below (37).

6 CONCLUSIONS

Bounds have been constructed on temperature and reactant concentration in the Semenov approximation (SA) (spatially homogeneous with reactant consumption) and in the complete problem (CP) (spatially distributed with reactant consumption).

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These bounds show that temperature and concentration are never negative and that the (dimensional) temperature in either case is bounded above by

$$EQc_0 e^{E/RT_0} / \rho C_V RT_0^2 \quad (44)$$

so long as activation energy is finite. The (dimensional) time constant for reactant consumption in either case ranges between

$$Ac_0^{m-1} e^{E/RT_0} f(c_0 Q E e^{E/RT_0} / \rho C_V RT_0^2)$$

and

$$Ac_0^{m-1} e^{E/RT_0}$$

The temperature bound (44) exceeds the adiabatic temperature $c_0 Q \delta C_v$, which is the maximum physically allowable temperature rise, because the approximate model (31-32) from which it was derived overestimates fuel supply. This is one example of a common occurrence with the bounding techniques used here; one can often identify the physical basis for a mathematically rigorous bound. Such a physical observation can then suggest a mathematical approach to improved bounds. For another example, see (40-43) and the next to the last paragraph of the Introduction.

In the exponential approximation ($\exp[\theta/(1+\epsilon\theta)] \approx e^\theta$), which corresponds to infinite activation energy in the kinetics term, temperature predicted by the models is not necessarily bounded. In this case, the (dimensional) induction period is no shorter than

$$-Ac_0^{m-1} e^{E/RT_0} \ln(1 - \rho C_V RT_0^2 EQc_0), \quad m = 1,$$

$$Ac_0^{m-1} e^{E/RT_0} (m-1)^{-1} / [(1 - \rho C_V RT_0^2 EQc_0)^{1-m} - 1], \quad m > 1.$$

Recalling our convention that references to infinite activation energy ($\epsilon \rightarrow 0$) are to the kinetics term alone, we note that infinite dimensionless temperature θ corresponds to infinite dimensional temperature T and conversely.

The temperature in the complete problem (CP), the spatially distributed reaction with reactant consumption, is always bounded above by the temperature field of the stationary approximation with finite activation energy, (FK _{ϵ}). This stationary temperature at finite activation energy is in turn bounded above by the temperature in Frank-Kemenetskii's stationary approximation with infinite activation energy, (FK₀). Consequently, one anticipates that critical behavior defined by sharp increases in temperature must occur in the

opposite order: the highest temperature reaction (FK₀) can exhibit critical behavior while the other reactions (FK _{ϵ}) and (CP) can still be subcritical at the same parameter values, and so on.

In particular, the critical parameter value δ_{crit} of Frank-Kemenetskii's infinite activation energy, stationary approximation appears to be a lower bound on analogously defined critical parameter values for both the finite activation energy, stationary approximation (FK _{ϵ}) and the complete, spatially distributed problem with reactant consumption (CP). This observation is based on the rigorous ordering derived above of the temperature fields in the three problems (FK₀), (FK _{ϵ}), and (CP), thereby providing an alternate view of the mechanism which orders critical parameters in the three cases. The three problems do not appear to have been considered simultaneously in this light by other investigators.

The results presented here do not depend on special restrictions on parameters or on approximations to nonlinear terms.

NOMENCLATURE

- A pre-exponential factor in Arrhenius kinetics term (mol/m³)^{1-m}/s
- B $EQc_0/\rho C_V RT_0^2$, dimensionless adiabatic temperature rise
- Bi $\chi d/\kappa$, Biot number
- c reactant concentration (mol/m³)
- c_0 initial reactant concentration (mol/m³)
- C_V reactant specific heat capacity at constant volume (J/K kg)
- d characteristic length of region enclosing reactant (m)
- D region enclosing reactant
- ∂D boundary of region enclosing reactant
- E activation energy (J/mol)
- $f(\theta)$ $\exp(\theta/(1+\epsilon\theta))$, dimensionless kinetics factor
- m reaction order ($m \geq 0$)
- Q exothermicity per mole (J/mol)
- R universal gas constant (J/K mol)
- S surface area of region enclosing reactant (m²)
- t^* time (s)
- t $t^* t_R = Ac_0^{m-1} t^* e^{E/RT_0}$, dimensionless time
- t_{ind}^* $t_R^* B / \rho C_V RT_0^2 e^{E/RT_0} EQAc_0^m$, (s) induction time for spatially uniform adiabatic reaction

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